

Length-dependent oscillations of the conductance through atomic chains: The importance of electronic correlations

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Abstract. – We calculate the conductance of atomic chains as a function of their length. Using the Density Matrix Renormalization Group algorithm for a many-body model which takes into account electron-electron interactions and the shape of the contacts between the chain and the leads, we show that length-dependent oscillations of the conductance whose period depends on the electron density in the chain can result from electron-electron scattering alone. The amplitude of these oscillations can increase with the length of the chain, in contrast to the result from approaches which neglect the interactions.

Quantum wires of atomic dimension are among the basic components of molecular electronics. The knowledge of the transport properties of such structures will be crucial for the design of devices, and the understanding of their properties is a major challenge in nano-electronics. Experimentally, chains of atoms between two electrodes have been formed and investigated [1–3], and the conductance of molecules like H₂ has been measured [4]. Besides the possible applications in molecular devices, this is of great general interest since electronic correlations alter considerably the properties of one-dimensional systems. Fermi liquid theory completely loses its applicability and Luttinger liquid [5] behavior appears. This has been observed for the case of metallic single wall carbon nanotubes [6]. A recent measurement on atomic Au, Pt and Ir wires between break junction contacts [7] has revealed oscillations of the conductance as a function of the length of the chain. For an odd number of atoms in the atomic wire the conductance has maxima while it is reduced when the number of atoms is even. This behavior is obtained after averaging over many realizations of the experiment in which the precise form of the contacts between the electrodes and the atomic chain might be different.

Since a few years, the conductance of chains of atoms has been the subject of an increasing theoretical activity (see Sect. 11.5.1 of Ref. [3] for an overview), and parity-oscillations had

been expected for chains of monovalent atoms like Au. The experimental observation of even-odd oscillations in chains of the polyvalent atoms Pt and Ir [7] however calls for more profound studies.

For a non-interacting half-filled tight-binding chain coupled to leads, Fabry-Perot like interferences lead to size-dependent parity oscillations in the conductance [8]. Taking into account the electron-electron interactions perturbatively [9] within the Hubbard model and using density functional theory for chains of Na atoms [10] indicate that these parity oscillations of the conductance persist once interactions are included in the model. Local charge neutrality has been identified [10–12] as the main cause of the even-odd oscillations in chains of monovalent atoms with a half-filled valence band. However, the phase of the parity oscillations depends on details of the model used to describe the coupling of the chain to the leads [11]. A four-atom period was obtained from density functional calculations for the conductance of atomic Al chains [13]. The analysis of the phase of these oscillations lead to the conclusion that the electron density in the chain is determined by the Fermi energy in the leads, and that local charge neutrality is not satisfied inside the chains.

Taking into account the full electronic correlations within a model of spinless fermions with next-neighbor interactions at half filling we showed that the even-odd oscillations can simply be the result of the presence of strong electronic correlations [14]. We even found that the interactions could enhance the amplitude of the parity oscillations in the conductance.

In this work we present a study of the length dependent oscillations in the conductance of atomic wires, taking fully into account the electronic correlations inside the chain. After presenting our numerical results for spinless correlated fermions in a one-dimensional chain, we will discuss the role of the shape of the contacts to the leads and the dependence of the conductance-oscillations on the particle density. These results will be contrasted with the theoretical predictions for models which neglect the interactions. This allows to identify signatures of the electronic correlations in the length dependent conductance oscillations which could be observed experimentally.

To calculate the conductance of a chain where electronic correlations may be strong, and which is coupled to leads, we consider the model Hamiltonian

$$H = -t \sum_i (c_i^\dagger c_{i-1} + c_{i-1}^\dagger c_i) + \sum_{i=2}^L U_i [n_{i-1} - V_+] [n_i - V_+] . \quad (1)$$

The first term describes the kinetic energy in the atomic chain (from site 1 to L) and in the leads. The operators c_i^\dagger create spinless fermions on the sites i of an infinite one-dimensional tight binding model, and the hopping element $t = 1$ is taken constant and sets the energy scale. The second term accounts for the interactions of strength U_i between particles on adjacent sites $i - 1$ and i inside the atomic chain. Here, $n_i = c_i^\dagger c_i$ is the density operator and the potential V_+ allows to adjust the particle density in the chain. A density $\nu = 1/2$ corresponding to half filling is obtained by $V_+ = 1/2$ which leads to particle-hole symmetry. To get other values for the density, V_+ is chosen in a self-consistent iterative way as described in Ref. [15]. Though the one-dimensional leads are of course not realistic, the suppression of the electronic correlations by the screening in real massive electrodes is nevertheless accounted for by the absence of interactions. The shrinking diameter of the contacts between the leads and the chain is modeled by a linear increase of the interaction strength U_i from zero to its full value $U_i = U$ over L_C sites [14].

To determine the conductance of the atomic chain in the presence of electronic correlations, we exploit the connection between its zero-temperature linear conductance and the persistent current or the charge stiffness of a non-interacting ring in which the interacting system is

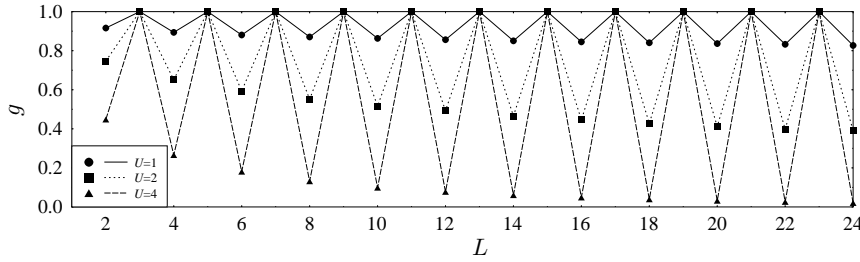


Fig. 1 – Conductance through interacting atomic chains at half filling ($\nu = 1/2$), as a function of their length L for the case of abrupt contacts ($L_C = 1$) and different values of the interaction strength.

embedded [14, 16, 17]. We use the density matrix renormalization group (DMRG) algorithm [18] to obtain the charge stiffness of the model (1) after closing it to a ring. For a detailed discussion of the method and the verification of its validity see Ref. [15] and references therein.

Results for the length-dependence of the dimensionless conductance g in units of the conductance quantum e^2/h are shown in Fig. 1 for the case of half filling. The presence of interactions leads to clear parity oscillations in the conductance through the atomic chain, with perfect conductance $g = 1$ for chains containing an odd number of sites, independent of the interaction strength. For even chains, the conductance is reduced when the interaction strength U increases. This reduction becomes more pronounced in longer chains.

In the limit of very strong interactions, the parity oscillations at half filling can be understood by considering the one-body hopping terms in (1) as a perturbation to the interaction term (for simplicity, we take abrupt contacts and $U_i = U$ for all $i = \{2, \dots, L\}$). In a chain of odd length L , the ground state of the interaction term alone is a superposition of two degenerate components. One of them corresponds to $N = (L + 1)/2$ particles occupying the odd sites $i = \{1, 3, \dots, L\}$, for the other $N - 1$ particles occupy the even sites of the chain. For infinitesimal hopping $t/U \ll 1$, the two degenerate components become coupled through sequences of N one-particle hopping processes. At half filling, when the Fermi energy in the non-interacting leads vanishes, this corresponds to a one-particle resonant tunneling situation. This results in perfect transmission and $g = 1$, independent of the weakness of the coupling between the effective bound state and the leads which is of the order of $t(t/U)^{N-1}$.

For the case of an interacting chain with even length L , the ground state in the absence of the hopping term of (1) is also a superposition of two components, $|\Psi_r\rangle = \prod_{n=1}^N c_{2n}^\dagger |0\rangle$ and $|\Psi_l\rangle = \prod_{n=1}^N c_{2n-1}^\dagger |0\rangle$. They correspond to the same number $N = L/2$ of particles, occupying the even and the odd sites of the chain, respectively. Even though these degenerate components are coupled by sequences of N one-particle hopping processes as in the odd case, these sequences do not involve particles hopping between the interacting chain and the leads. Therefore, these processes do not contribute directly to the transport through the chain. The process which dominates zero-temperature transport through the chain in the even case is coherent cotunneling. Taking into account the lowest order processes in t/U , involving transitions *via* N -particle states with modified charge configuration and states having $N \pm 1$ particles in the chain leads to a decrease $\sim U^{-L}$ of the conductance with the interaction strength U for even chain length L . This is a signature of the Mott insulating behavior occurring for infinitely long chains at $U > 2$. The strong suppression of the even conductance with the length at $U = 4$ in Fig. 1 can be understood as a precursor of the insulating behavior.

In the limit of weak interactions, we can calculate the conductance as $g = 4 |G_{L,1}(0)|^2$,

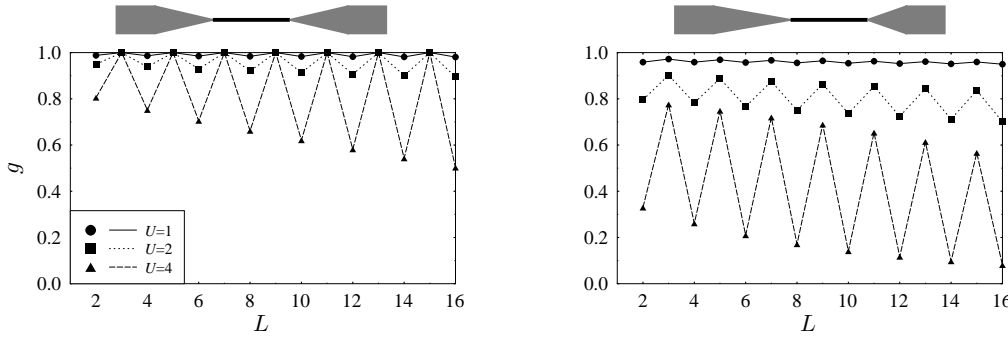


Fig. 2 – Conductance as a function of the number of sites in the interacting region for $\nu = 1/2$ and symmetric contacts of length $L_C = 3$ (left), and asymmetric contacts of length $L_{Cl} = 4$, $L_{Cr} = 2$ (right). The geometries we have in mind are sketched above the graphs.

where $G_{L,1}(0)$ is the Green's function for the propagation of a particle from the first to the last site of the chain at the Fermi energy of the leads $E_F = 0$, within the full many-body Hamiltonian [9,19], evaluated at zero temperature. We can easily calculate this Green's function in second order perturbation theory in U . The conductance of odd chains is not affected by this perturbation, but in the case of even chains it is reduced by an amount proportional to U^2 , consistent with the numerical results for small chains and weak interaction [14]. Such an approach has been used to show that weakly interacting Hubbard chains connected to non-interacting leads exhibit parity oscillations [9].

We now address the role of the shape of the contacts between the atomic chain and the leads in order to consider the more realistic case of smooth contacts. The conductance for a symmetric setup with a chain connected to the leads through two contacts of length $L_C = 3$ is shown in Fig. 2 (left), at half filling. The even-odd oscillations as a function of the chain length persist in the presence of such smooth contacts, and the perfect transmission of odd chains is not affected. However, the reduction of the conductance in even chains is less pronounced when the contacts are smooth and the amplitude of the oscillations is smaller. Nevertheless, the dependence of the oscillations on the interaction strength and on the length of the chain are similar to the situation of abrupt contacts. Only in the unrealistic limit of infinitely smooth contacts one expects perfect perfect transmission for even chains as for the odd ones [14] and the parity oscillations disappear.

While it seems reasonable to assume symmetric contacts when describing certain experimental situations like break junctions, it is difficult to exclude the case of a slightly asymmetric situation where the two ends of the atomic chain are connected to the electrodes in different ways. This breaking of the reflection symmetry of the model has consequences that can be seen in Fig. 2 (right). Here, the left and right contacts are assumed to be of length $L_{Cl} = 4$ and $L_{Cr} = 2$, respectively. Most strikingly, the conductance obtained for odd chains no longer reaches the unitary limit $g = 1$ and decreases with increasing interaction strength and chain length. As compared to the symmetric situation, the conductance values are reduced for the even chains as well. However, the parity oscillations persist and the phase of the oscillations is robust as well: one has maxima of the conductance for odd and minima for even chains.

The parity oscillations that we have studied so far occur in the case of half filling which corresponds to chains of monovalent atoms with a half-filled valence band like Na and Au. At other values of the filling, the period of the length-dependent oscillations in the conductance is

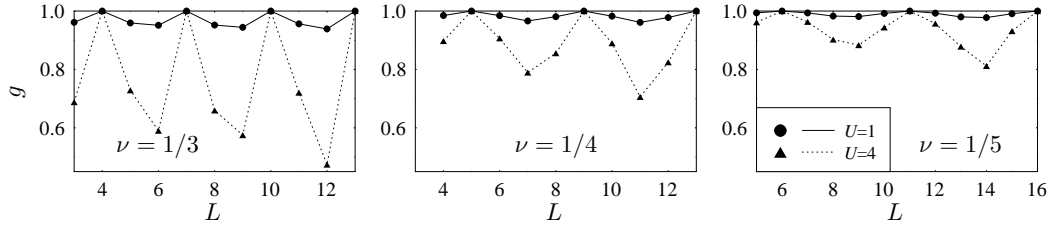


Fig. 3 – Conductance as a function of the length of an atomic chain connected to the leads by abrupt contacts for fillings $\nu = 1/3$ (left), $\nu = 1/4$ (center), and $\nu = 1/5$ (right).

different. To illustrate its dependence on the filling, we show in Fig. 3 results for the densities $\nu = 1/3, 1/4$, and $1/5$. For simplicity, we use abrupt and symmetric contacts. In this case, perfect transmission appears for chain lengths of $L = 1 + q/\nu$ with integer $q = \{0, 1, \dots\}$, and the period of the length-dependent oscillations is $1/\nu$ sites for $\nu < 1/2$. At larger fillings, the density of holes $1 - \nu$ is relevant for the periodicity of the conductance. Furthermore, it can be seen that the amplitude of the oscillations is smaller when the filling is lower. This can be understood from the fact that the conductance oscillations in our model are caused by the short range electron-electron interactions. At low filling, this interaction becomes less relevant since the probability to find two particles on adjacent sites of the chain decreases.

Setting the interactions to zero ($U = 0$) in our model leads to a perfect homogeneous chain which exhibits perfect transmission and conductance $g = 1$ for all fillings and lengths L , and the length-dependence of the conductance disappears. This shows that electronic correlations alone can be the origin of length-dependent conductance oscillations in atomic chains. In a non-interacting model, another scattering mechanism is necessary for reproducing the oscillating behavior of the conductance. A possibility is to suppose that the contacts between the atomic chain and the electrodes are a source of reflections.

This can be achieved by adding the contact term

$$H_C = (t - v_l) (c_1^\dagger c_0 + c_0^\dagger c_1) + (t - v_r) (c_{L+1}^\dagger c_L + c_L^\dagger c_{L+1}) \quad (2)$$

to our Hamiltonian (1). This replaces the hopping matrix elements t between the chain and the left and right leads by the couplings v_l and v_r , respectively. Choosing $|v_l|, |v_r| < t$, this describes a chain of L atoms coupled to one-dimensional leads by weak links which give rise to Fabry-Perot like interferences. In this model, the particle density in the chain is the same as the one in the leads which is determined by the Fermi energy. This situation is different from assuming local charge neutrality as discussed in Ref. [13]. Using a transfer matrix formalism we can calculate the transmission at the Fermi energy, and using Landauer's formula $g = |t(k_F)|^2$ the conductance

$$g = 16 \sin^4 k_F \left| (v_l - v_l^{-1}) (v_r - v_r^{-1}) e^{2ik_F L} - (v_r e^{2ik_F} - v_r^{-1}) (v_l - [v_l e^{2ik_F}]^{-1}) \right|^{-2} \quad (3)$$

of the non-interacting system with the Fermi wave number k_F . This result is symmetric with respect to the exchange of v_r and v_l .

The only L -dependence of g in (3) is through the factor $e^{2ik_F L}$. Therefore, the result is periodic in L with period π/k_F . In dimension one, the Fermi wave number is related to the filling through $k_F = \pi\nu$, and therefore the period of the oscillations is given by $1/\nu$ sites as in the interacting case discussed above. However, the phase is different, except for half-filling,

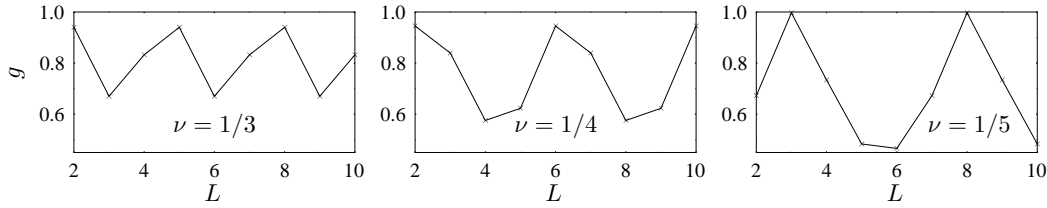


Fig. 4 – Conductance calculated from Eq. (3) as a function of the number of sites in a non-interacting chain which is connected to one-dimensional leads through weak links of strength $v_l = v_r = 0.75t$. The filling is $\nu = 1/3$ (left), $1/4$ (center), and $1/5$ (right).

where one obtains always even-odd oscillations with the maxima for odd N . This can be seen from Fig. 4, where the analytical result of Eq. (3) is shown for the example of $v_l = v_r = 0.75$ and different fillings. Assuming local charge neutrality and using a tight-binding model with a filling which is independent of the filling of the leads, the same periods have been found in the conditions for resonant transport [12]. However, the phase of the oscillations is different [13].

As compared to the interacting case, the behavior of the non-interacting model presents striking differences. First of all, without interactions the length-dependence is perfectly periodic and the conductance satisfies $g(L + 1/\nu) = g(L)$. Thus, the amplitude of the oscillations is independent of L . This is because the motion of the particles between the contacts is ballistic. In contrast, for interacting particles, the length-dependent oscillations we obtained (see Fig. 1) are superimposed with a decrease of the conductance minima and an increase of the amplitude of the oscillations with the length of the chain. For asymmetric contacts, the amplitude remains more or less constant, but the conductance decreases with the length for both, maxima and minima of the oscillations (see Fig. 2). While such a decrease is not seen in the experimental data for Au chains, Pt and Ir do show [7] such a behavior which is reminiscent of our results for correlated chains, but difficult to explain within non-interacting models.

In addition, while the maxima in our correlated model always reach perfect conductance $g = 1$ in the case of symmetric coupling to the leads, without interactions this is the case at half filling and only for special parameter combinations outside half filling [12]. In the case of asymmetric contacts $v_l \neq v_r$ the amplitudes of the length-dependent oscillations obtained from the conductance of Eq. 3 are somewhat reduced, but remain clearly observable, similar to the case of weakly correlated chains in Fig. 2. This is consistent with the fact that the oscillations have been observed experimentally [7] in a situation where the contacts were very probably not exactly symmetric.

The period of the length-dependent oscillations predicted by both non-interacting approaches agrees with the result of our numerical calculations for a correlated chain. This indicates that the period is indeed determined by the filling of the conductance band alone, and given by $1/\nu$, independent of details like the shape of the contacts between the chain and the leads. However, outside half filling, the phase of the oscillations is different for all three mentioned approaches. Therefore, we can expect that, in contrast to the period, the phase depends on fine details of the experimental situation. As a consequence, a more refined analysis than averaging over many realizations of the preparation of the chain as done in Ref. [7] could be necessary for their observation.

In conclusion, we have studied the effect of electronic correlations on the length-dependent oscillations observed in atomic wires. In the absence of interactions, these oscillations can

be understood as an interference effect between two scatterers connecting the chain to the leads. However, the electrons of an atomic chain form a correlated system which can by itself be an important source of scattering, and hence of resistance. We have studied the consequences of this additional scattering mechanism within a non-perturbative many-body approach, and have shown that it leads to very similar conductance oscillations. This suggests that electronic correlations are an additional and independent reason for their occurrence. In the presence of reflection symmetry, this mechanism leads to perfect conductance at the maxima of the oscillations, for all fillings. The length-dependent oscillations persist in the presence of asymmetric contacts between the chain and the leads. The period of the oscillations is given by the inverse of the conductance band filling, independently of the model and the mechanism leading to the oscillations. But the phase of the oscillations depends on details of the model and whether or not interactions are present. In non-interacting models, the electrons are mainly scattered in the contacts, and the conductance oscillations are periodic. Electronic correlations lead to the occurrence of scattering processes in the whole chain and result in a decrease of the conductance with the length, in addition to the oscillations. This is a striking difference between our correlated chain and non-interacting models. As a consequence, the amplitude of the oscillations depends on the length, and can even increase with the length of the chain when electronic correlations are present. These findings should be relevant for distinguishing in the interpretation of recent and future experiments on the length-dependence of the conductance of atomic chains and molecular wires the many body effects coming from electron correlations from the effects describable by simpler one body theories.

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